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Acoustic detector for fission neutrons

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Abstract

We report here on the progress towards the development of an acoustic neutron detector. The goal is to make a detector that is passive, low cost, and scalable to large areas. We will discuss the potential for this detector to be blind to gamma radiation and the potential for high-efficiency fast neutron imaging. The detector signals are based on the generation of either bubbles or thermo-acoustic shock in a liquid due to energy deposition from a neutron. The interaction of a neutron with a hydrogen rich fluid will primarily be a neutron-hydrogen collision. As they are nearly identical in mass, the most probable scattering event is an elastic event, depositing approximately half of the collision energy into the hydrogen atom. For fission neutrons this energy is in the 1-15 MeV range. The excited hydrogen atom (proton) rapidly thermalizes with the fluid establishing a thermo-acoustic signature which we then monitor with a hydrophone submerged in the fluid. Further, if the fluid is near a phase transition, the deposition on energy from the neutron can form a bubble. The formation and collapse of the bubble will create a shockwave which is then detected using a hydrophone submerged in the fluid. Here we will describe: the generation and propagation of acoustic-shock waves, the design and construction of an acoustic detection cell, and the callibration of the cell using photo-acoustic spectroscopy. We will discuss tests preformed using both cosmogenic neutrons and a Californium 252 source, and conclude with a discussion on future implementation of the acoustic neutron detector.

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Summary

New technique for the passive detection of neutron based on thermo-acoustic shock waves. The shockwave comes from the interaction of a neutron with a hydrogen atom in the fluid and is detected with a commercial hydrophone. The detector consists of a hydrogen-rich fluid (diethyl-ether, isopropanol, acetone...) in a container and a hydrophone. The goal of the LDRD is to demonstrate, characterize, and quantify the acoustic signals generated from the interactions of neutrons with hydrogen rich fluids. Finally, due to the nature of the neutron interaction for shockwave formation, the detector is predicted to by blind to gamma radiation.

1 Introduction

A long studied technique for detecting high-energy particles is the well known bubble or cloud chambers. These are systems that are heated or pressurized beyond the point of phase change in such a fashion that the system can spontaneously undergo that phase transition if disturbed. The preparation of the fluid into the supercritical state leaves the system in a meta-stable configuration that is highly unstable to perturbations. When a high energy particle or ionizing radiation interacts with the system, this unstable equilibrium is disrupted causing a localized phase change. This takes the form of droplet formation in a cloud chamber and bubble formation in a bubble chamber. The underlying interaction is energy transfer from the particle into the supercritical system. The deposited energy locally disrupts the equilibrium leading to the phase change. If the properties of the fluid are sufficient, and enough energy is deposited, a bubble is nucleated. The nucleated bubble can cascade into multiple bubbles and even causing the entire system to undergo a phase change.

In the early-1950's bubble chambers were coupled with sonic transducers, or microphones, to detect the nucleation of the first bubble [3] as a way to trigger the recording the data acquisition for particle generation events. This gave rise to the first uses of acoustic transducers, or microphones, to be used for particle detection.

In the 1960's several groups began to work on tensioned fluid systems [7, 5]. These are fluids that have some type of transducer driving an acoustic wave or vortex. At the nodes, the fluid is above its supercritical point and can be used as a particle detector, just like a bubble chamber. The effect was so dramatic that it was reported that upon interaction with a high energy particle, bubbles form and rapidly collapse resulting in "an audible noise and the release of light" [5]. In these experiments, standard microphones where used, outside the chamber, to record the events. The experiments were able to detect individual neutrons. The drawback to these detectors was the limited active area, set by the size of the chamber, the frequency, and the amplitude of the compression wave.

Another set of neutron detection experiments utilized superheated droplets suspended in gels, the detectors became known as SDDs or superheated droplet detectors [1, 2]. In the SDDs, droplets of supercritical fluids are isolated in some form of a matrix gel to keep the droplets stable. Interaction of a neutron with a droplet would cause bubble formation, and in the proper gels, the resultant bubble is held in the matrix gel until they are redout. This makes these SDDs excellent for neutron dosemetry [8]. They are a commercially available product, and have been adapted for either direct readout of counting using an acoustic pick-up.

In the research presented here, we are proposing an acoustic detector for neutrons that requires higher energy deposition then the previously reported experiments. Higher energy particle detectors using acoustic signatures have been proposed for neutrino detection.

In the search for understanding the energy distribution of cosmogenic neutrinos, the detection of extremely high energy (GeV) neutrinos is relevant [4]. In these detectors, large tanks of water were used as the detector material. While GeV neutrinos have weak interactions with the water,

they can deposit substantial energy, through particle showers, leading to acoustic shockwave formation and bubble formation. Predictions are that both of these processes can then be detected using a hydrophone and give complementary information to Cherenkov detectors. Currently, there are several large scale neutrino detectors that are working on incorporating arrays of hydrophones to detect and triangulate neutrino interactions. The most prominent is the international detector in the Mediterranean sea, off the coast of France known as the ANTARES project, with the acoustic detection being the AMADEUS detector subproject [4, 6].

In this LDRD we are looking for thermo-acoustic shock waves and/or bubble formation due to the deposition of a few MeV on energy into a near supercritical fluid, at STP (standard temperature and pressure) from single and multiple neutron interactions. In section 2 we will discuss signal formation and propagation, energy deposition, and sample selection. In section 3 we will discuss the design and construction of the acoustic cell and the properties of the acoustic detectors, and in section 4 we will discuss the calibration of the system using photo-acoustic spectroscopy. In section 5 we will discuss our progress toward neutron detection and finish with a discussion on future directions in section 6.

2 Signals generation

The thermo-acoustic signal is a signal generated due to localized heat deposition into a fluid. This problem was originally studied in the early 1950's and an analytic function was derived for the time scale, t_{ta}, of the resulting acoustic signature, such that $t_{ta} = \frac{L}{c}$, where L is the length over which the heat is deposited and c is the speed of light. This formalism assumes that the heat is deposited quickly compared to all other timescales in the system. Thus the relevant time scale is the fastest a particle can move, the speed of light. The response to such a impulse gives rise to a pressure gradient ΔP, which then propagates following a the inhomogenious wave equation;

$$\Delta P - \frac{1\partial^2 P}{c_s^2 \partial t^2} = -\frac{\alpha}{C_p}\frac{\partial q(r,t)}{\partial t^2} \tag{1}$$

where C_p is the heat capacity of the fluid, α is the coefficient of heat expansion of the fluid, and $q(r,t)$ is the energy distribution of the deposited heat. The solution to the equation is an impulse wave that propagates, in a dispersionless system, to the detector.

In practice, the signals are generated in finite systems with both dispersion and walls. The dispersion gives rise to a decrease in amplitude and the wall interactions cause reflection which can either constructively interfere or destructively interfere, depending on the geometry of the container, position of the detector and location of the signal generation.

The maximum energy that a neutron, with energy E_N, can deposited into an atom with atomic number A is given by;

$$E_{max} = 4\frac{AE_N}{(A+1)^2} \tag{2}$$

Thus the maximum energy a neutron, with energy E_N, hitting a hydrogen atom can transfer is E_N. Although, the most probable energy transfer is an elastic collision where $\sim 1/2E_N$ is deposited in the hydrogen atom and the neutron leaves with $1/2E_N$ of energy. In these systems, we are concerned with energy deposition in the order of a few MeV into the fluid over a distance of the hydrogen atom recoil or a few nano-meters. The smallest length scale is proton diameter on the femto-meter scale, so crudely we are interested in acoustic frequencies between $\sim$ 300Hz and 300kHz.

The fluids of interest are those that are nearly at a critical phase change at room temperature, thus depositing MeV of energy could result in localized phase change, and acoustic shock, and bubble formation. For instance Diethyl ether boils at 34.6 degrees C, just 14 degrees above room temperature, while methanol boils at 65 degrees C. For the purpose of acoustic neutron detection Diethyl ether is a preferred candidate to methanol. A goal of the current research is to investigate the acoustic neutron signal in different hydrogen rich fluids and explore the energy dependance of the signal size.

One other potential for the generation of the thermo-acoustic signal is localized chemistry initiated by the neutron interaction with the fluid. The scenario is that a neutron collides with

a hydrogen atom and abstracts the hydrogen from its parent molecule. Ether, C2H10O, consists of two pairs of CH3-CH2 linked by an oxygen, or CH3-CH2-O-CH2-CH3. Upon abstraction of a hydrogen atom, the molecule becomes an unstable radical with a peroxide group at its core, capable of undergoing further exothermic reduction, releasing chemical heat into the system. While ether may not be the ideal compound for this process, there is a potential for chemical amplification of the thermo-acoustic signal.

3 Apparatus

The apparatus is designed to be a focused acoustic system. The design is such that an acoustic wave originating anywhere inside the chamber will be focused onto the signal microphone. The criteria for the cell design are that it have a significant mass such that low frequency vibrational noise does not couple into the acoustic cell and interfere with the signal. We designed in optical access for calibration of the acoustic signals using photo-acoustic spectroscopy, which requires optical access for the laser beam. The cell is also equipped with an overflow valve and a pressure release valve to ensure both proper fill of the vessel and proper ventilation of the solvents used.

3.1 Acoustic cell

The design of the acoustic cell is a simple stainless steel cube with a parabolic surface machined into the bottom of the cell to focus all acoustic waves onto the holder for the microphone. Figure 1 shows the cross-sectional machine drawings of the cell design. The outer structure is a heavy stainless cube 10 cm by 10 cm in inner diameter and weighing a little over 20 lbs. There are two recessed window groves machined into the side of the cube for optical access. Two, two inch BK7 windows fit into the groves and are sealed to the cell with o-rings. The windows are held in place with Delrin holders that bolt to chamber in a manner as to minimize stress on the glass.

We have designed a special holder for B&K commercial microphones to hold the microphone such that the surface of the microphone will be wetted by the detector fluid under the maximum fill condition of the cell, and also provide electrical and signal connections to the microphone. The main fill hole is plugged with 3/8-inch Swagelok plug.

Figure 2 shows the competed cell filled with the detection fluid. As will be discussed later, the B&K microphone the chamber was designed for, is not compatible with the volatile solvents we are using for detector fluids. Thus the image show the hydrophone that is now used as the primary detector in the system.

3.2 Microphones and Hydrophones

The main proposal was to use a gas phase microphone that would be in tensioned contact with the detection fluid to collect the acoustic signal. This turned out to be a challenge as the volatile fluids have a tenancy to migrate across the thin stretched metalic membrane and condense inside the gas phase microphone, shorting out the microphone. The gas phase microphone membrane is a stretched thin nickel film around a superstructure with an electrode placed microns from the surface. This combination forms a capacitor. Vibrations are read out as a transient in the capacitor current. We used several of the 4000 series microphones from B&K with a biased pre-amplifier EG&G 402, with a 100V bias. The individual microphone assembly is shown in figure 3. This was the setup used for the gas-phase methanol calibration discussed in section 4. To date, all attempts to look at liquid phase signal with the detector have not been successful.

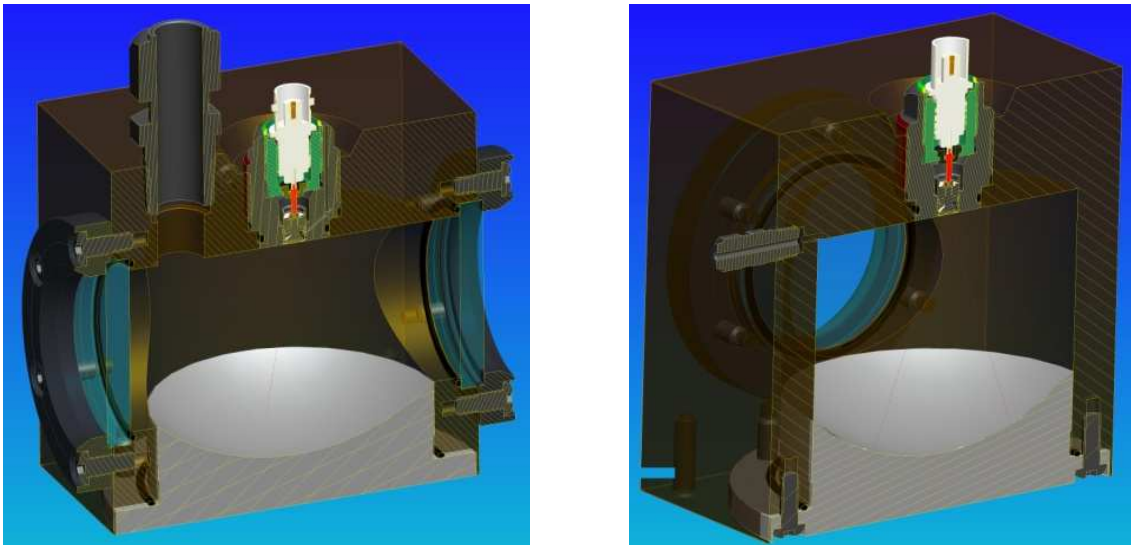

Figure 1. Machine drawings and cross-sections for the acoustic cell. The windows are for calibrating the cell with photo-acoustic spectroscopy, while the holder on top is a compression fit connector for the microphone.

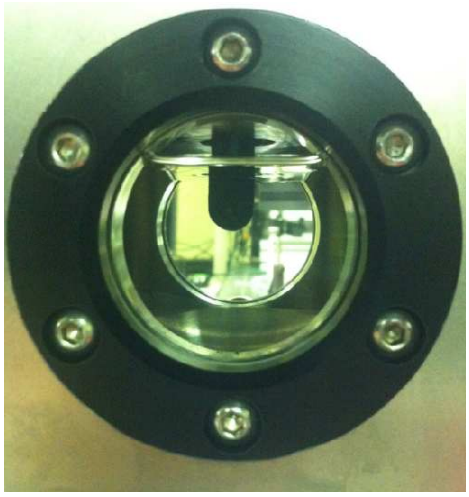

Figure 2. A photograph of the completed cell with a hydrophone and filled with methanol.

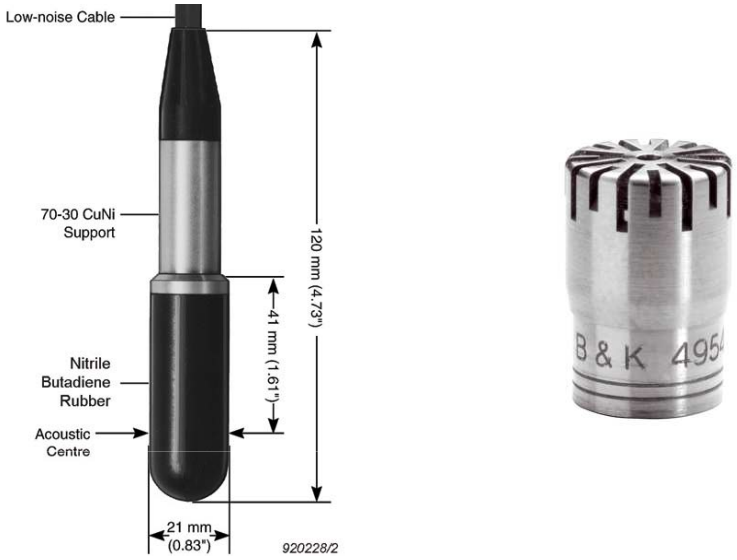

Figure 3. Stock photos of a commercial B&K hydrophone and microphone. The left image is shows a hydrophone assembly. The active area is embedded in Nitrile for protection. The right image shows a standard B&K stretched nickel microphone with protection cap. These images were taken from *www.bksv.com*.

For the liquid phase measurements, both the calibration and the neutron detection, we employed a B&K hydrophone, model 8004. The sensor is piezo-electric acoustic sensor, and as a result is nearly a factor of 1000 less sensitive then the gas phase microphone. However, the transducer is coated in Nitrile to handle wet and harsh environments. The transducer can operate with or without biasing. In practice we find that the bias voltages improve the response frequency range and signal. We used 100V, similar to the gas phase microphone with the same pre-amplifier system. The left image of figure 3 shows the hydrophone used along with the dimensions.

The exact specifications of the acoustic transducers; such as frequency response and sensitivity, depend on the mode of operating, termination of the electrical signal and cable length between the pre-amplifier. As we are not to the point in this research where an ideal operational mode has been chosen, the specific values will not be given.

4 Calibration

The first step in the operation of the acoustic neutron detector is to characterize the response time scale and amplitude for a known perturbation. For this we have chosen to perform photo-acoustic spectroscopy on the overtone of methanol. Methanol was chosen as it is a potential fluid for the detector and has similar properties to the other potential detector fluids. For instance, the speed of sound in methanol is 1143 m/s, while the speed of sound is 1174 m/s for acetone and 985 m/s for ether. This makes methanol a good surrogate for signal propagation in the volatile detector fluids we are considering. Further, methanol overtone spectra have been well studied allowing us to verify our results with literature. The importance of actually taking a spectrum in the fluid is to distinguish between background acoustic signals generated either in the room or from the laser hitting dirt or impurities and not from energy deposition into chemical bonds. We specifically want the signals generated by depositing energy into chemical bonds as this best mimics the thermo-acoustic signals expected from the neutrons.

4.1 Photo-acoustic data

We performed three sets of calibration experiments; the first is a gas phase methanol spectrum and signal, the second is a liquid phase spectrum and the final set of experiments was to investigate the arrival time of the acoustic signal as a function of where the probe laser was focused inside the cell. The experimental setup was to focus a 600nm laser beam into the cell through a f = 300mm lens such that the focus was in the middle of the cell, underneath the microphone. The laser light was generated from a 5ns seeded Nd:YAG laser pumping a linear dye laser with LD610 dye. The oscillator was tweaked using a Kiton620 dye such that the laser could scan between 590nm and 620nm to ensure a flat intensity over the 595-610nm range. Figure 4 shows a liquid phase acoustic signal and it's reflection off the cell wall. To collect spectrum we placed an Ortec box-car integrator over the initial rising slope. The laser was run at a repetition rate of 30Hz and the signal was integrated for 1s. The laser was then scanned at a rate of 0.01nm/s and the spectra recorded using a 16bit National Instruments USB-6211 interface.

Figure 5 (a) shows the gas phase spectra of methanol on the (0-4) overtone. The features match the reported spectra found in literature, and demonstrates that the acoustic signals are being generated from energy deposition into the vibrational motion of the methanol. Based on the reported cross-sections of $\sim 2\ 10^{-23}$ cm^2 and the energy per pules from the laser of 10μJ, we estimate that $\sim$ 100keV of energy is being dumped into the methanol, giving rise to the detectable acoustic signal.

Figure 5 (b) shows the calibration curve for liquid methanol. The high collision rate in the fluid, compared to the background, rapidly quenches the excited vibrational state, drastically broadening out the features such that all we observe is a broad featureless spectrum. While this spectra was not found in literature, it is consistent with other liquid phase spectrum, in that the transition from gas phase to liquid phase smears out the spectrum and shifts the center frequency.

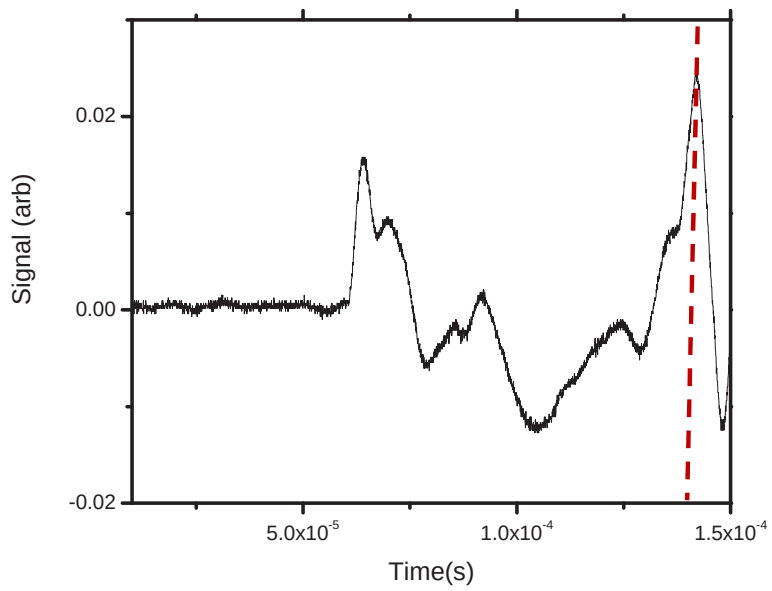

Figure 4. This figure shows a typical large amplitude photo-acoustic signal. The dominant reflection shows up at the dotted red line.

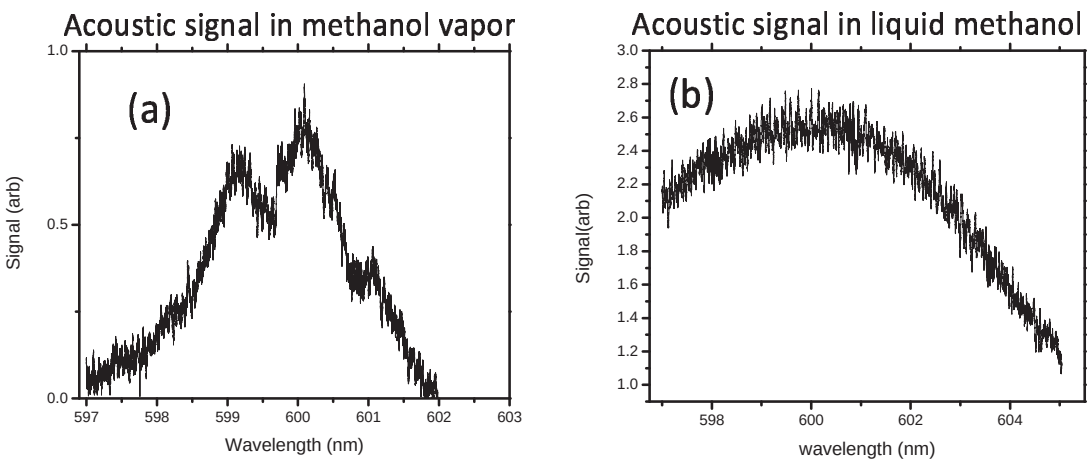

Figure 5. Photo-acoustic spectrum of methanol. Spectra (a) is the photo-acoustic spectra of the $0 \leftrightarrow 4$ overtone in gas-phase methanol near 600nm. The data was recorded using the B&K stretched nickel microphone. (b) is the photo-acoustic spectrum of liquid-phase methanol taken with a B&K hydrophone near 600nm.

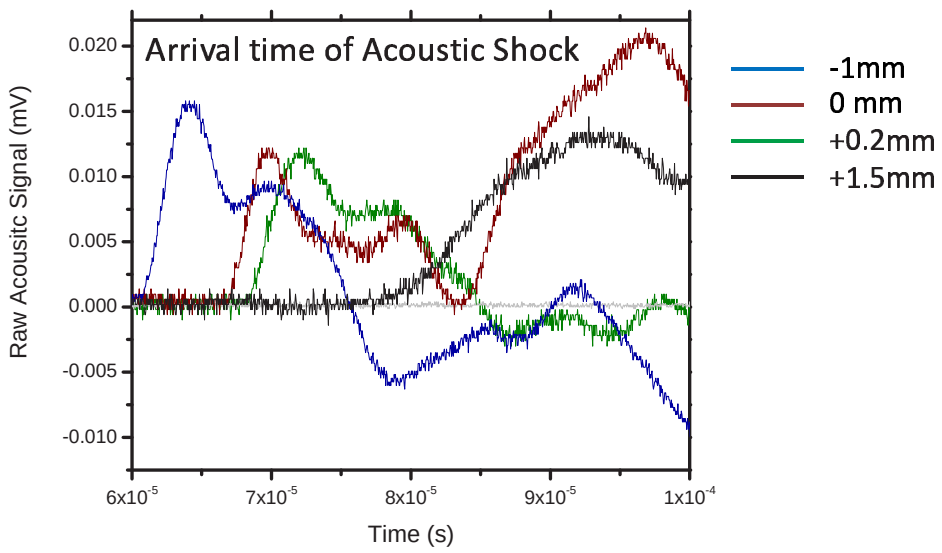

Figure 6. Arrival time variation for the leading edge of the acoustic shockwave. The laser beam focus was translated from its initial position, marked 0mm, and the arrival times were recorded with respect to the laser pulse.

Figure 6 shows the leading edge of the acoustic signal in liquid methanol for a series experiments where the laser power and frequency are kept constant while the vertical position of the laser focus is varied. The arrival time of the signals are all timed to the laser beam trigger. What is observed is that a translation of 200μm can clearly be decerned by eye, and that 50 μm of translation can be distinguished from a single acoustic detector. Using multiple detectors we predict that signal location can be determined to within $\sim$ 10 μm.

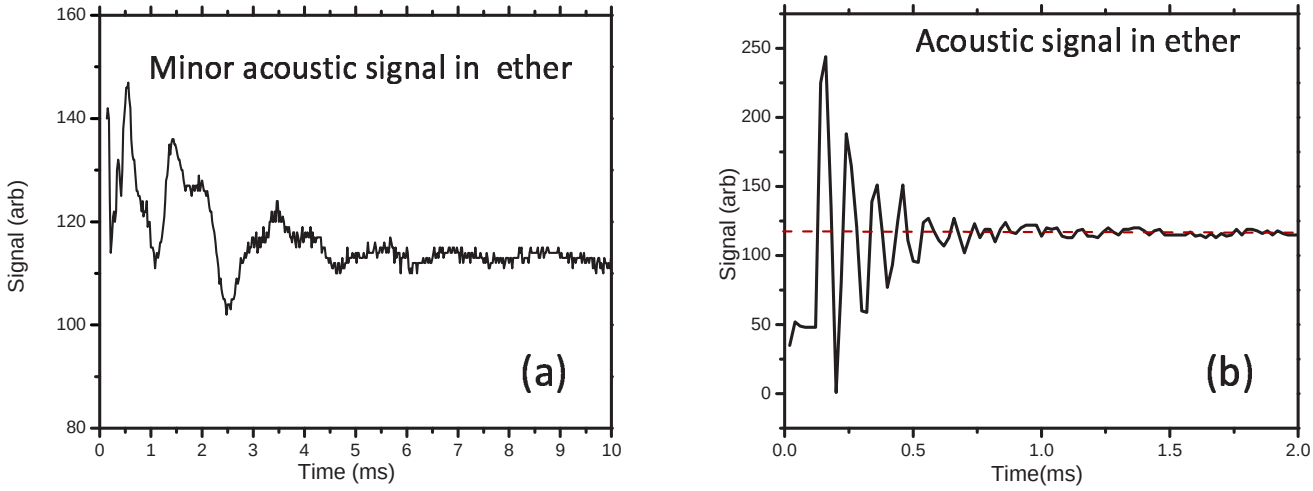

Figure 7. (a) shows a minor, low amplitude long-frequency event, while (b) shows a large amplitude moderate frequency event. The major events were observed at a rate of about one per hour while the minor events were on the order of one per 20 minutes.

5 Neutron detection

For the first set of neutron detection experiments we set up the system to look for background, or neutrons created in the upper atmosphere during particle showers. The estimated rate is one neutron every 10 to 20 seconds passes through our cell, but most of the neutrons are at thermal energies and not predicted to give a thermo-acoustic signal. In the experimental setup we determined a reasonable threshold level to discriminate between random acoustic noise and the predicted signal. If the detection scope triggers, the data is streamed to a computer and stored. Then the scope is reset to collection mode. We ran the experiment during the nights when there was minimal activity in the laboratory building.

Two sets of experiments were performed: one in liquid methanol and the other in diethyl ether. There were no statistically signifcant event with an acoustic signal structure (ringing) in any of the 10 methanol runs, for a combine data acquisition time of 120 hours. Figure 7 shows some of the data collected in the runs with Diethyl ether. There were 3 runs, with a total data acquisition time of 36 hours. We observed two characteristic signal, the major and minor signals. The minor events, figure 7 (a) were recorded at a rate of 1 per 20 minutes and major events, figure 7 (b), at a rate of about 1 event per hour. While the observation rates are low, the comparison with the methanol data leads to the conclusion that the recorded events may be due to neutron events. However, there are many possible scenarios for the generation of these signals from unknown noise sources. While these results are quite promising, they are not conclusive evidence for acoustic detection of neutrons.

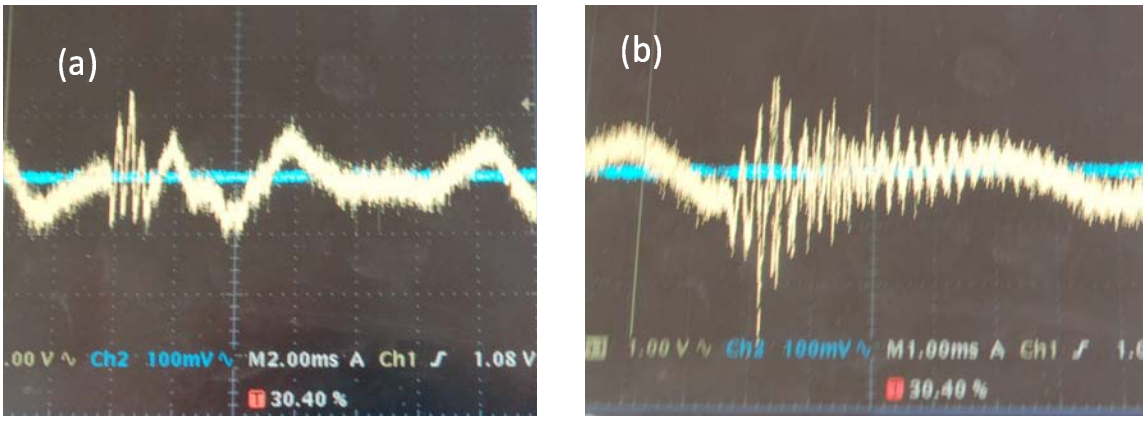

Figure 8. Two acoustic signals recorded in the presence of a Cf-252 neutron source.

The final set of experiments that were completed with in the time frame of this LDRD was to introduce a neutron source near the detector. For this we used a relatively hot Californium 252 source with a measured activity of 60 μCi peaked at 1.5MeV. Figure 8 shows two typical signals. The large variation in the baseline is due to some type of large noise source in the pre-amplifier system. As yet we do not understand the origin of this noise and are currently working on identifying its origin. The signals were collected at a rate of 1 every 20 seconds. We would anticipate about 20/second based on the source's activity and proximity to the detector. When the source was removed the signal count rate dropped significantly but was still none-zero. No major events, like the signals in figure 8, were observed when the detector was filled with methanol or just empty. If the signals are due to neutrons, the rate would be consistent with observing neutrons at a energy of 8.5 MeV and up, based on the detection rate. As a result, we can not conclusively say that the observed signals are from neutron interaction events. However, like the background neutron signals, these signals look extremely promising but they are not conclusive as we are still waiting on a statistical event and not triggering of a neutron event. Repeating these experiments with a triggered neutron source is the next experimental step.

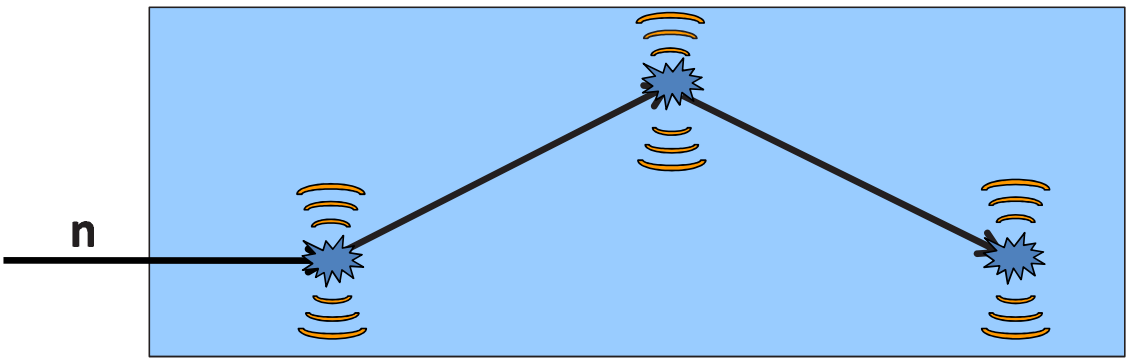

Figure 9. A conceptual drawing for a multi-event detector. Placing multiple hydrophones around a cell large enough for multiple events, and monitoring the arrival time signature of the events, the acoustic detection technique can be used to triangulate the individual interactions and monitor the neutron trajectory for imaging or source location.

6 Conclusion

The data we have collected is very suggestive of the production and detection of an acoustic signature from neutron interactions in a passive fluid. The results suggest that methanol is to tightly bound at room temperature for the production of the acoustic signature, while diethyl ether is bound weak enough to be sensitive to neutrons with energies greater than 8.5MeV. Further, the arrival times of the acoustic signals can be used to identify where in the passive detector the signal was generated. Combining these results we speculate on a future passive neutron detector illustrated in figure 9 where a single neutron enters the passive detector that is surrounded by acoustic detectors, each interaction gives rise to an acoustic signal which is triangulated to give the flight path of the neutron, yielding a high efficiency neutron “camera” for source location and fast neutron imaging.

References

[1] RE Apfel. Superheated drop detector. *Nuclear Instrumentation Methods*, 162:603–608, 1979.

[2] T Biro, A Kelemen, and I Pavlicsek. Acoustic detection of neutrons by bubble detectors. *Nuclear Tracks and Radiation Measurments*, 17:587–589, 1990.

[3] DA Glaser. Some effects of ionizing radiation on the formation of bubbles in liquids. *Physical Review*, 87:655, 1952.

[4] K Graf, J Hobl, U Kappes, R Lahmann, C Naumann, and K Salomon. Towards acoustic detection of uhe neutrinos in the mediterranean sea- the amadeus project in antares. *Journal of Physics: Conference Series*, 60:296–299, 2007.

[5] B Hahn and RN Peacock. Ultrasonic cavitation induced by neutrons. *Nuovo Cimmento*, 28:334–340, 1963.

[6] R Lahmann. Status and first results of the acoustic detection test system amadeus. *Nuclear Instruments and Methods in Physics Research Section A*, 604:S158–S163, 2008.

[7] D Lieberman. Radiation induced cavitation. *Physical Fluids*, 2:466–468, 1959.

[8] G Size, G Zhang, N Banhfa, Z Changjun, G Yongjing, C Zhe, X Caijin, L Chao, and L Cunxiong. Acoustic response of superheated droplet detectors to neutrons. *Nuclear Instruments and Methods in Physics Research A*, 668:21–25, 2012.

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